DESIGN AND FABRICATION OF PERMEABILITY APPARATUS FOR DETERMINATION OF PERMEABILITY OF POROUS CERAMICS

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By
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CERTIFICATE

This is certified that the work contained in the project entitled "DESIGN AND FABRICATION OF PERMEABILITY APPARATUS FOR DETERMINATION OF PERMEABILITY OF POROUS CERAMICS" by Rajeev Hansdah (Roll no-109CR0648) in partial fulfillment of the requirements of the award of Bachelor of Technology Degree in Ceramic Engineering at the National Institute of Technology, Rourkela is an authentic work carried out by him under my supervision and guidance. To the best of my knowledge, the matter embodied in the thesis has not been submitted to any other university / institute for the award of any Degree or Diploma.

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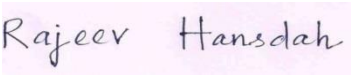

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ABSTRACT

Permeability measurement determines the pore size and pore connectivity is an important characterization for porous materials. For the measurement of permeability highly sophisticated & expensive instrument are commercially available for research purpose but for undergraduate level research the purchase of this equipment is an expensive affair though the working principle of the apparatus is very simple. So an attempt has been made to design and fabricate a permeability measurement set up for undergraduate level research work at very low cost using Darcy's equation as basic principle. Initial sketches and designs were drawn manually on the basis of text book diagrams and research papers. Subsequently the designs were analysed with respect to ease of fabrication, cost and adaptability to various size of samples. Finally the final design was selected out of 3 designs and the three dimensional design of the final design was made using CATIA (version 5) software, after which the equipment was fabricated using brass. Brass is used for its easy machinability and long life. The measured permeability showed similar trend for similar sample measured by other equipment (Mercury porosimetry).

CHAPTER-1: INTRODUCTION

1.1 **Introduction:**

Permeability is a most important material property which defines the resistance to the passage of the fluid, and it depends on the porosity and pore connectivity. Therefore the permeability is correlated with the type of pores and pore size distribution. The permeability is determined from Darcy's equation. The permeability measurement gives an idea of the packing behaviour of the particles in the body. The correct measurement of permeability is very important in many physics and engineering fields such as soil science, particulate systems, reactive reactor medium, fabrics, porous ceramic and filter processing and their applications.

The usual apparatus for permeability measurements essentially contain a set up where a fluid is forced to pass through a porous medium.

1.2 The guiding Principle-Darcy's equation [1]:

This semi empirical equation describes fluid transport in porous media based on one or more fluid.

$Q= (K_p \Delta P A)/L\eta$

Q=volume flow rate

K_p= specific permeability coefficient

ΔP= pressure drop across the sample

L=flow length or thickness of the test sample

A=area of cross-section of the sample

η= fluid viscosity

The permeability coefficient K_p depends on the combination of the fluid and porous material used. The greater the value of K_p the higher will be the rate of flow of a fluid through a material [1].

CHAPTER-2:
LITERATURE REVIEW

2.1. Permeability and its applications:

The measurement of permeability is of great important in broad range of fields which follow the principles of porous media physics

- In hydrology permeability has great important it helps in studies of infiltration, redistribution of water in the unsaturated zone, recharge of aquifers and groundwater flow within aquifers [2].
- In the petroleum industry the permeability of porous rock with oil filling is an important area where measurement of permeability is used [2].
- In environmental engineering the permeability of soil, sediments, rocks, clay and artificial porous media is great important in determining toxic waste disposal facilities, the main aim here is to separate the waste to greatest extent possible [2].
- In agriculture for the study of the transport of water in and near the root zone, related to the problems of irrigation and management of soil the measurement of permeability is important [3].
- In soil mechanics the permeability is important which determine soil water content at a given time and place which influence the rate of soil consolidation and the stability of infrastructure[3].
- In medical application, permeability is used for determining drug delivery system [3].

For measurement of permeability there are highly sophisticated and expensive apparatus are their commercially available for the above mentioned applications and for research purpose. So purchase of this instrument for undergraduate level research work is an expensive matter though working principle is very simple. In commercially available permeability apparatus hydraulic pressure is used to force the fluid through a define volume of the porous medium. Some of the commercially available permeability measurement equipment is shown in Figure 2.1 and Figure 2.2

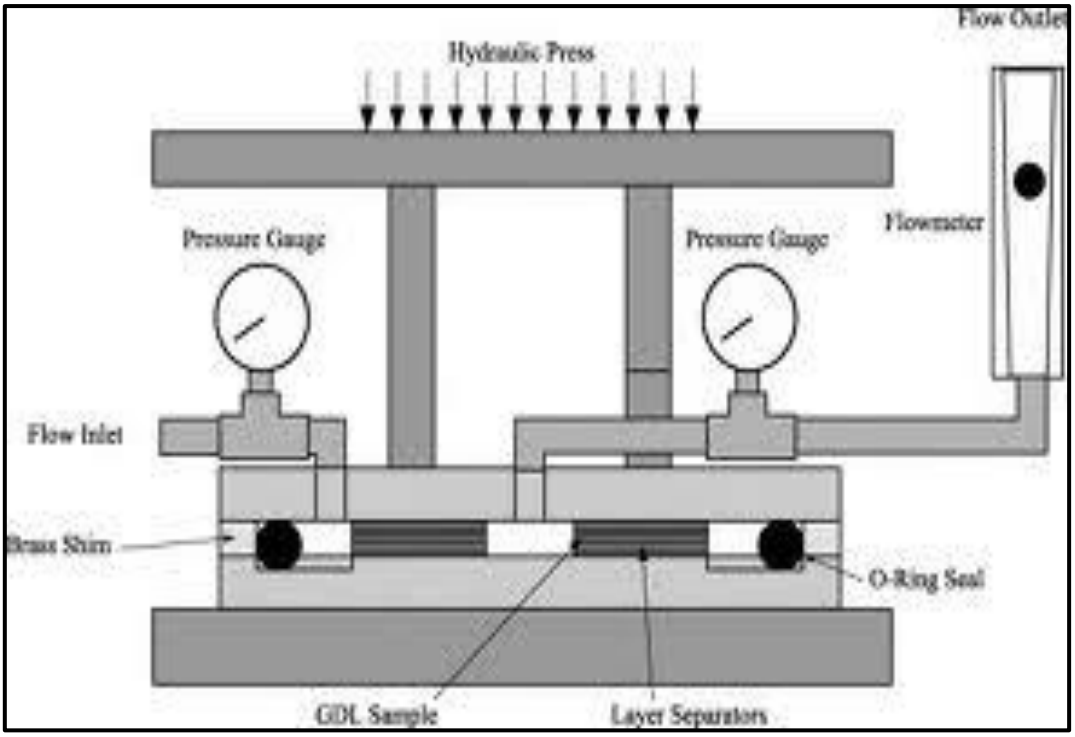

Fig. 2.1 Commercially available permeability measurement instrument [4]

Fig. 2.2 Commercially available permeability instrument [4]

2.2 Objective and novelty of work:

- To design and fabricate a simple and low cost permeability measurement set up which can be used for undergraduate level research work.
- Use of water head pressure to force the liquid though porous sample.
- To measure the permeability of porous samples prepared in the laboratory.

CHAPTER-3:

DESIGN OF APPARATUS

Initially three designs were under consideration. Out of the three designs finally only one design (or its modification) was finalized. The following section provides the different aspect of the three designs.

3.1 Design-1:

The design is shown in Figure-3.1.The design consists of

(a) Two metal mesh support between which sample will be placed and pressure will be applied.

(b) Two rubber seal would be provided on the outer wall of the mesh for sealing the gap, to prevent leakage of liquid at the time of applying pressure.

(c) A filter medium to filter the liquid

(d) A balance to measure the weight of water permeated in a specific time for knowing the volume flow rate of liquid.

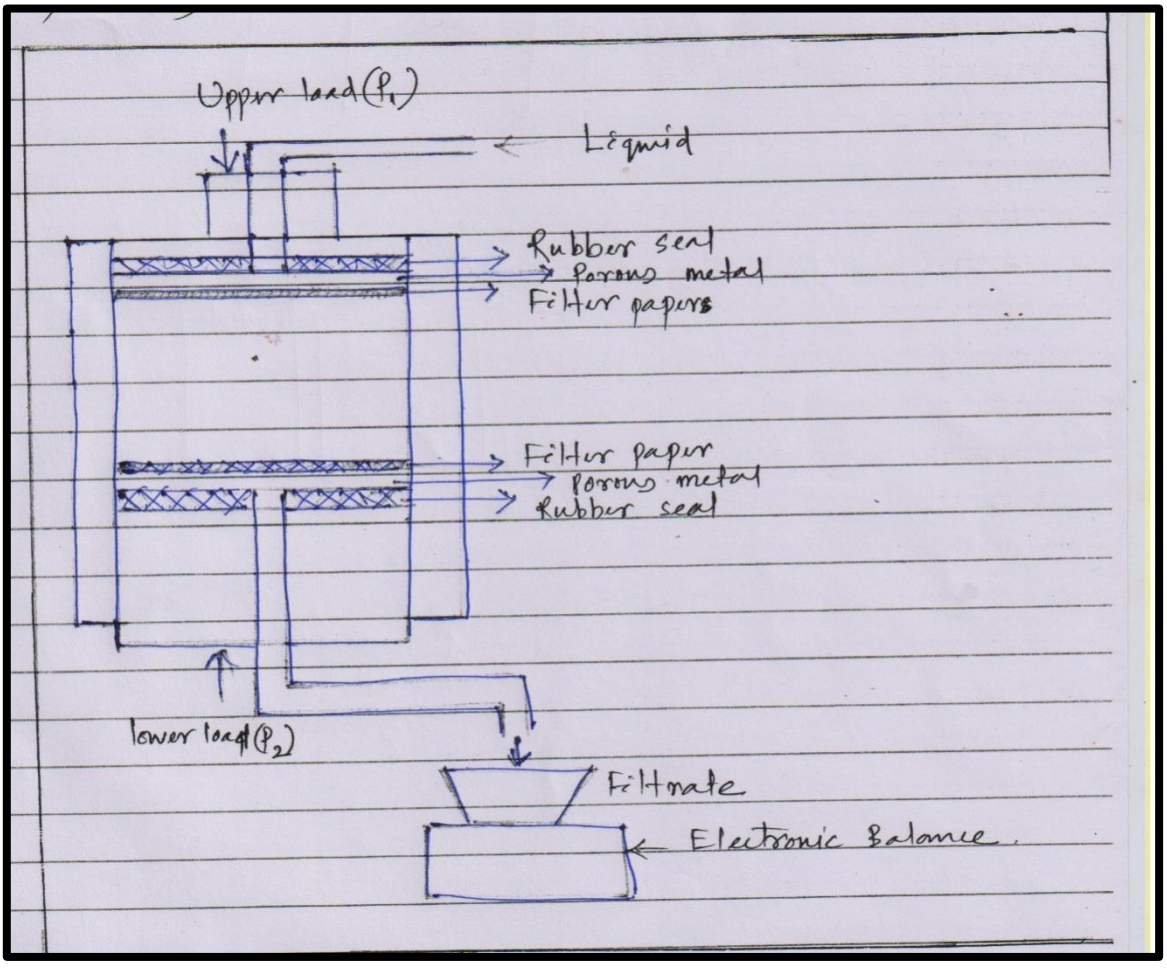

Fig. 3.1 Design-1 of permeability measurement apparatus

The pressure is applied at the top and bottom part is at atmospheric pressure, so ΔP is the difference between the applied pressure and atmospheric pressure. Therefore, for measurement of permeability the design of apparatus should have facility for measurement of volume flow rate as well as pressure drop across the sample.

It was observed that in this design it is difficult to measure the pressure accurately. The main aim was to design a simple model which could be fabricated easily and where porous sample prepared by different shape forming methods (pellet pressing method, gel casting method) could be used. Therefore, further modification of Design-1 was considered and this is discussed in Design-2.

3.2 Design-2:

In the second design, a laboratory bench press is considered as pressure source and an electronic balance is arranged at the bottom of the collector vessel for measuring the volume flow rate of liquid(Figure- 3.2).

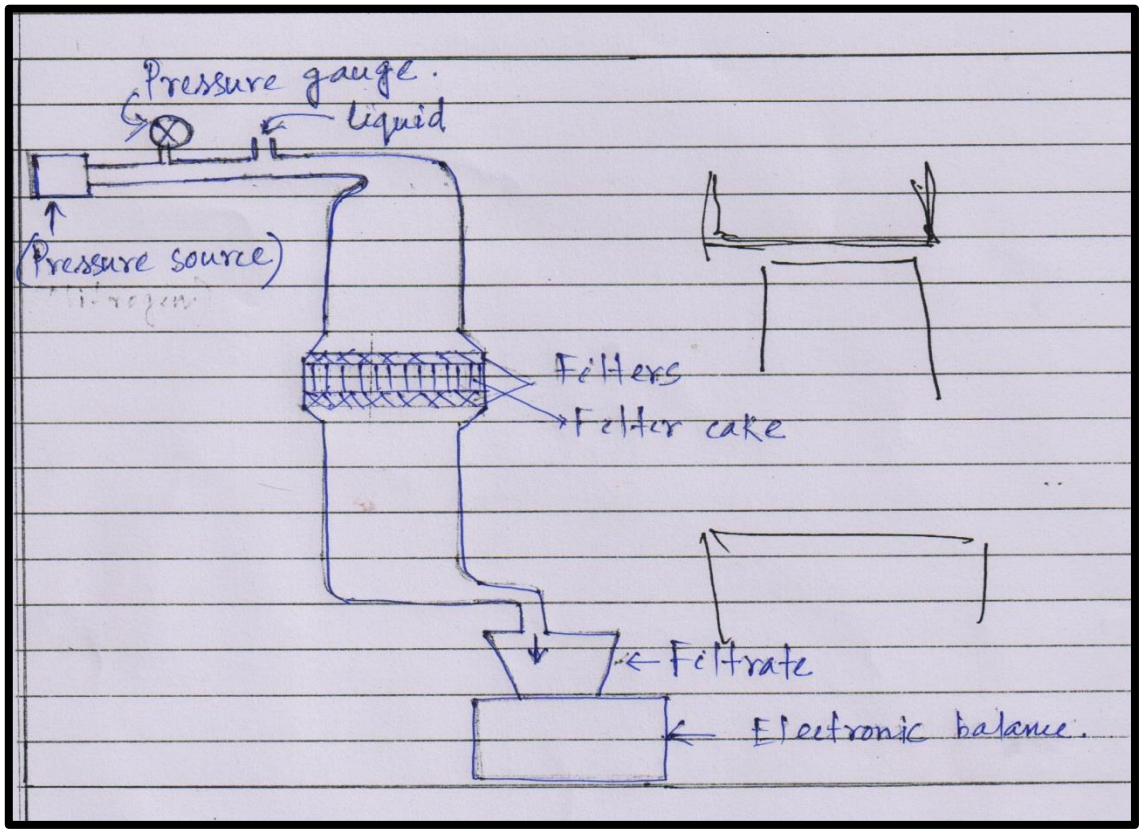

Fig. 3.2 Design-2 of the permeability measurement apparatus

The sample is placed in the sample holder and the liquid is passed through sample holder .Filtration will start when pressurized gas (air or nitrogen) is applied from the above. The filtrate liquid is collected to calculate the volume flow rate.

This design was also not tried further because of the strict requirement on sample specification as well as the availability of gas for pressing. However from the design it was realized that an adaptable sample holder need to be prepared which will be flexible enough to hold all kind of samples prepared by different methods. The incorporation of sample adaptability led to development of Design-3.

3.3 Design-3:

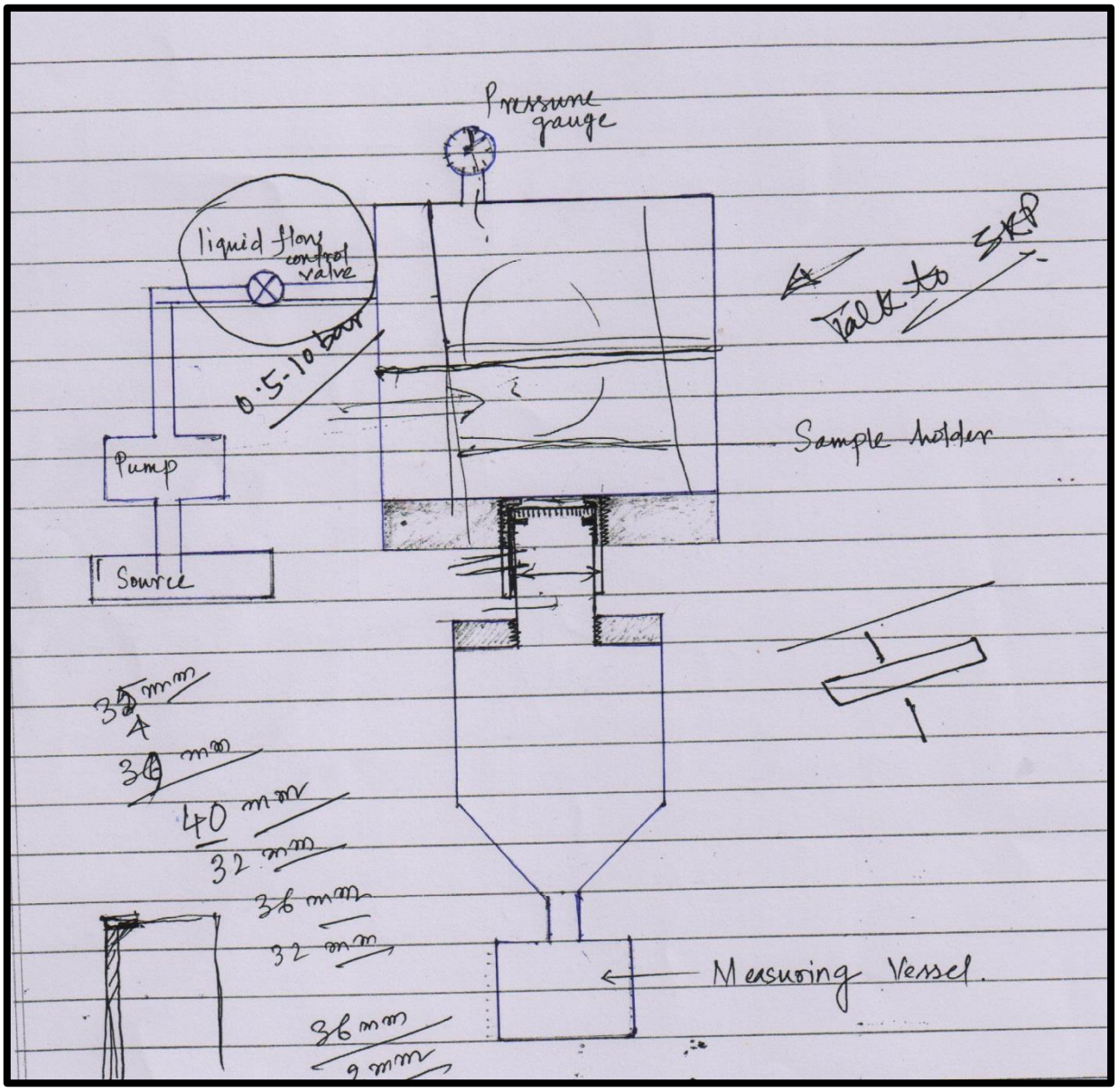

Fig. 3.3 Design-3 of the permeability measurement apparatus

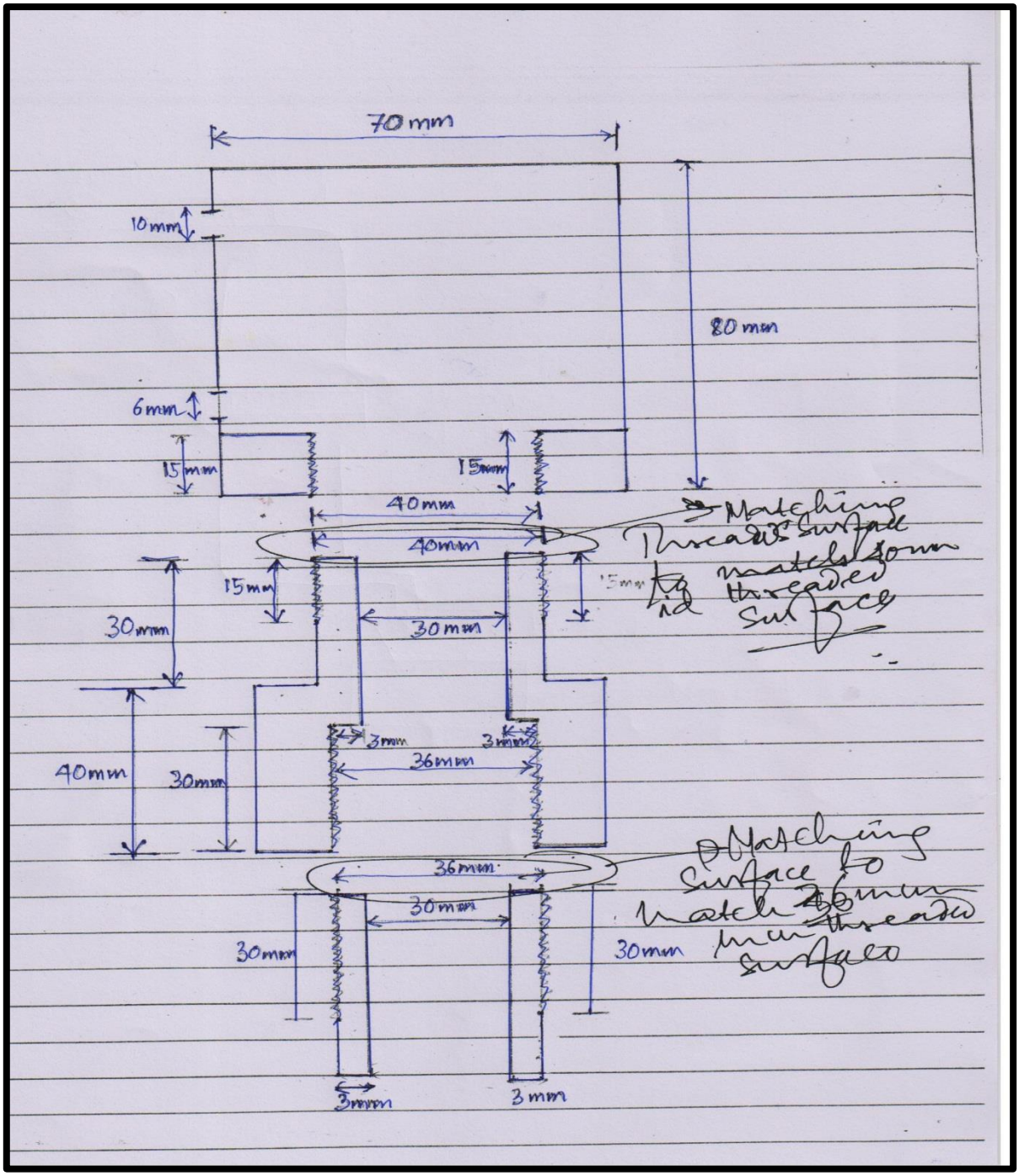

Fig. 3.4 The final design of the permeability apparatus

The initial design is show in the Figure-3.3and final design is shown in Figure-3.4. The final design consists of tank for holding liquid (water), sample holder, pump, liquid flow control valve, pressure gauge and measuring cylinder. The design is comparatively simpler and fabrication will be easy. Here the, main point is the preparation of sample holder .The sample holder plays most important role because accuracy of flow rate depends on how accurately and tightly the sample is held by

sample holder ensuring that there is no leakage of liquid through sides. So the sample holder was separated from the tank. Further the use of detachable sample holder also provided a means of using different sample with different holder while keeping the tank and other arrangement same.

3.4 Three dimensional designs:

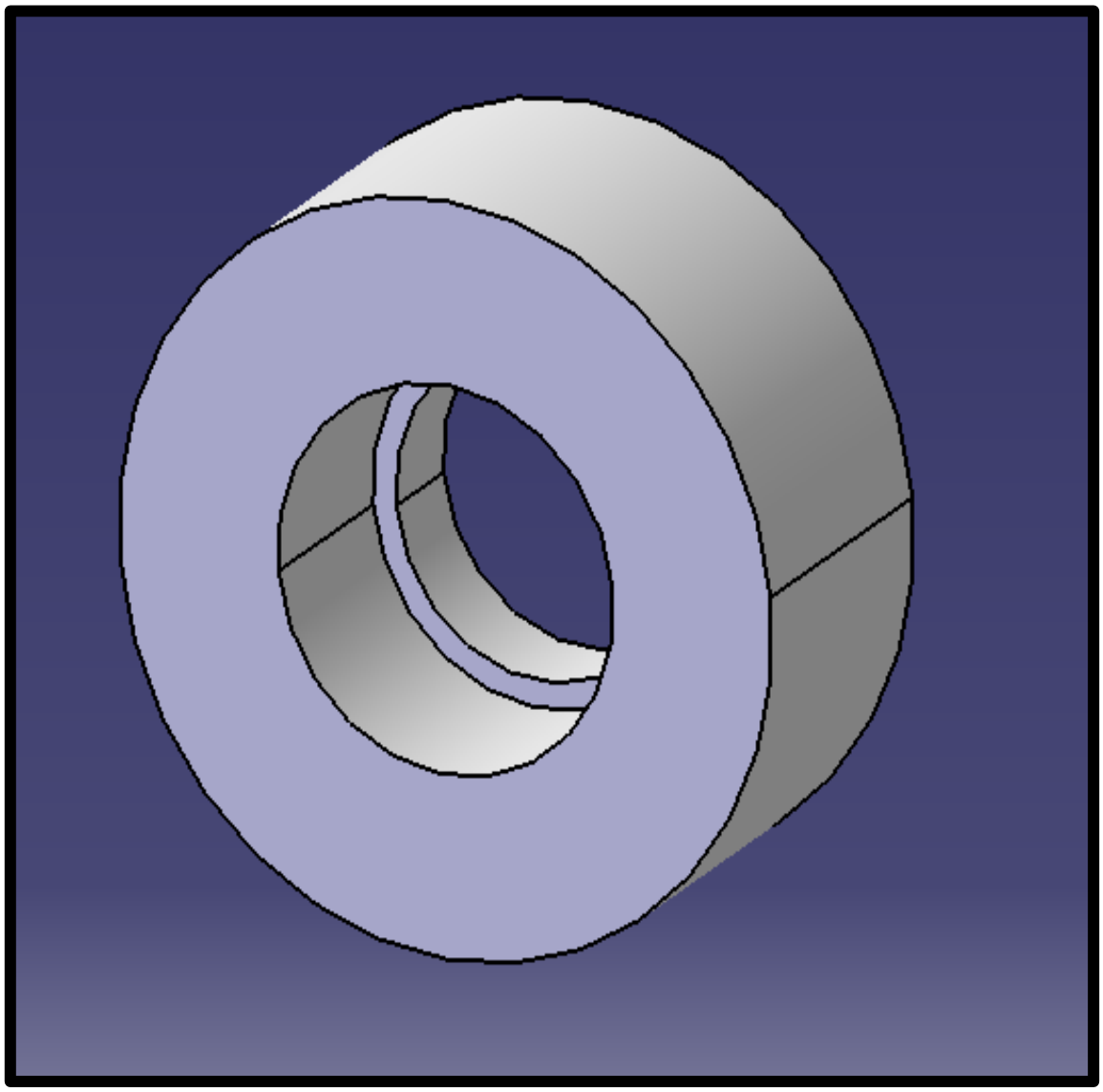

Fig. 3.5 Isometric view of the sample holder

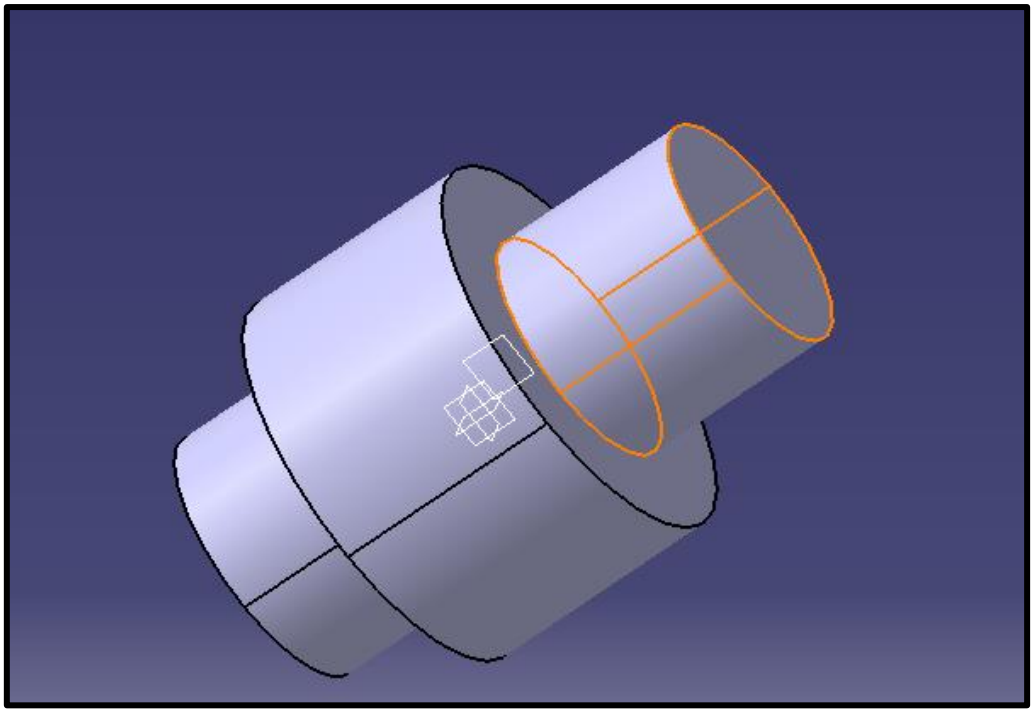

Fig. 3.6 Isometric view of sample holder with bolt

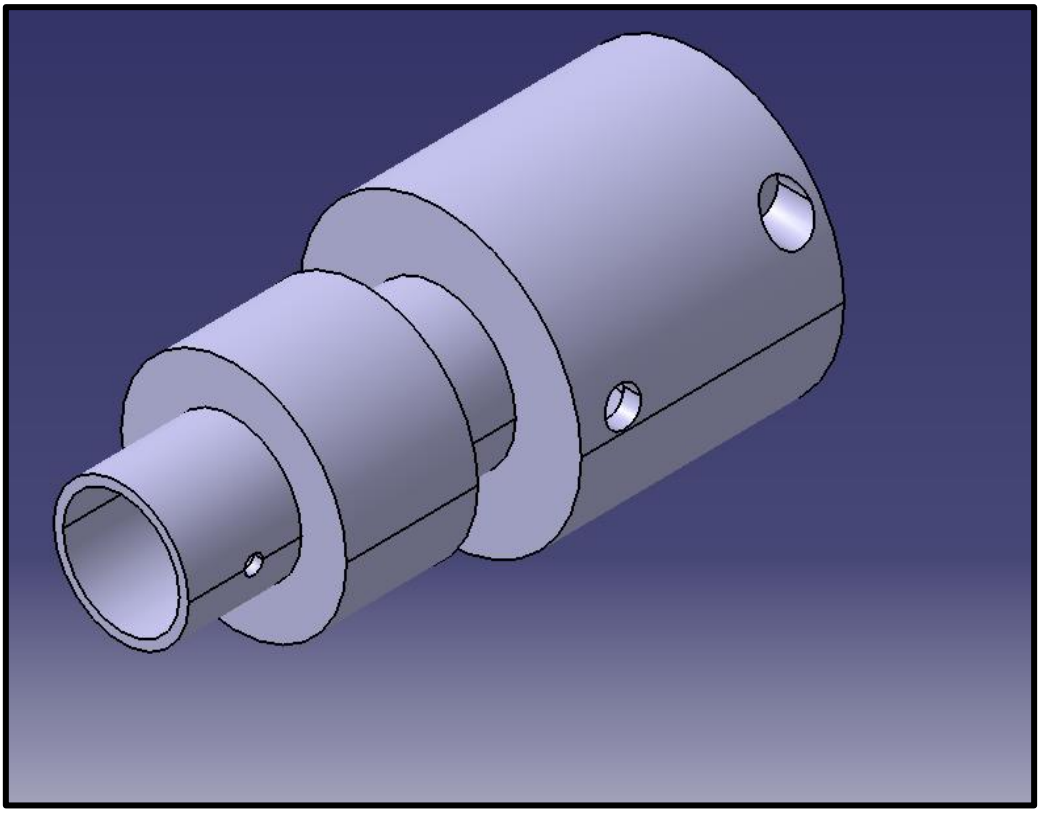

Fig. 3.7 Isometric view of the final design

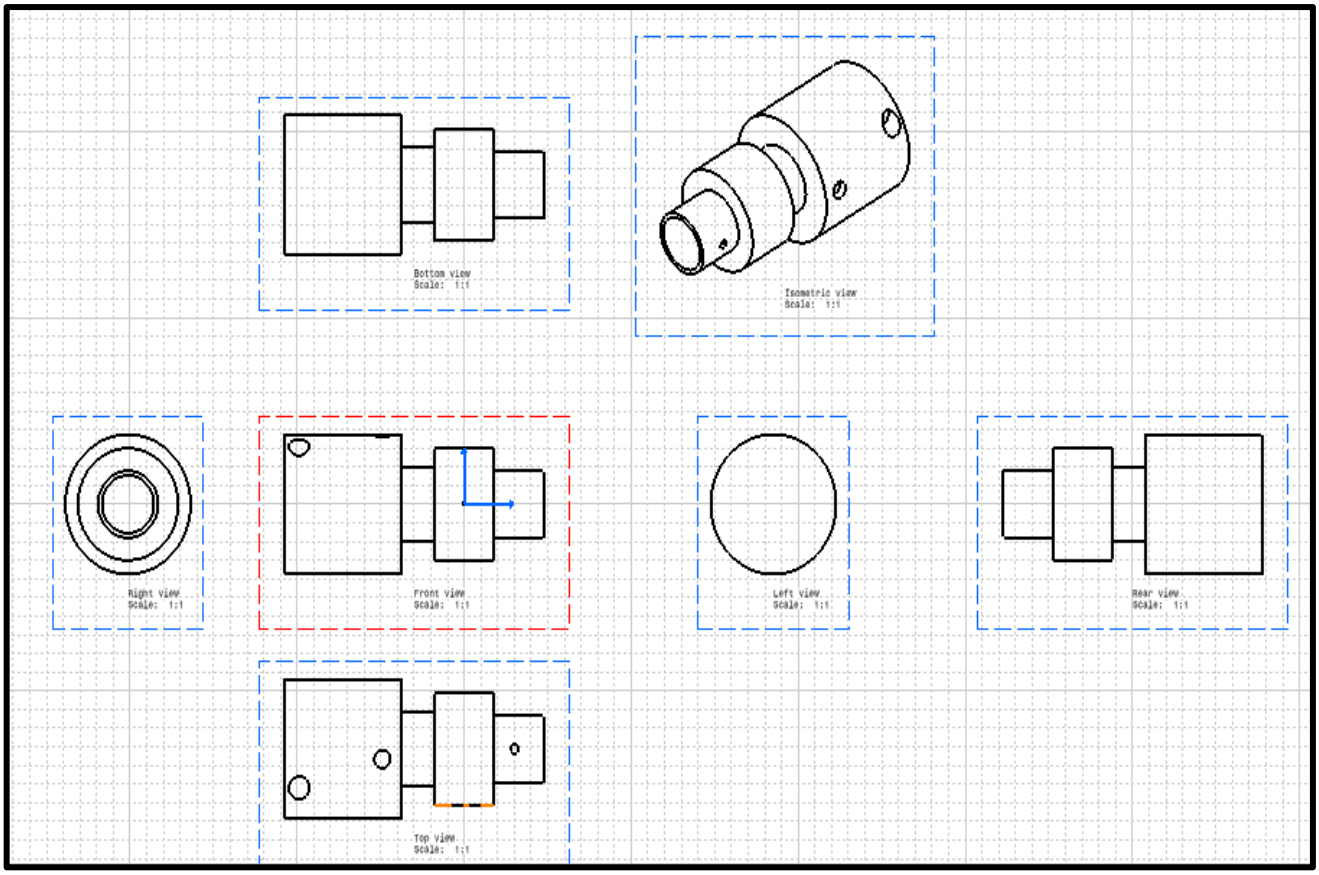

Fig.3.8 Sectional view of final design.

CHAPTER-4:

FABRICATION OF APPARATUS

4.1 Fabrication:

The fabrication of apparatus was done according to final design. It was fabricated with brass because brass is easily machinable and it has long life.

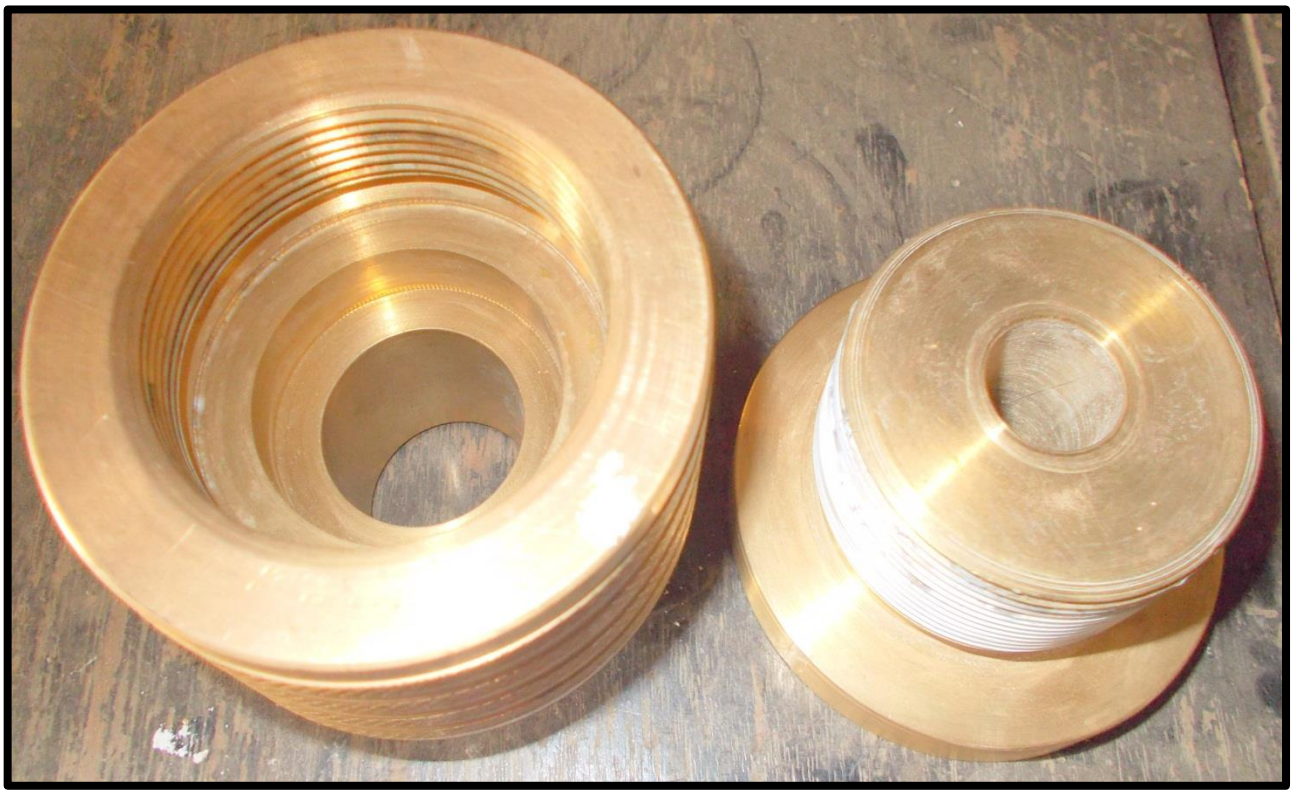

Fig. 4.1 Picture of sample holder (left) and bolt (right) to tight the sample

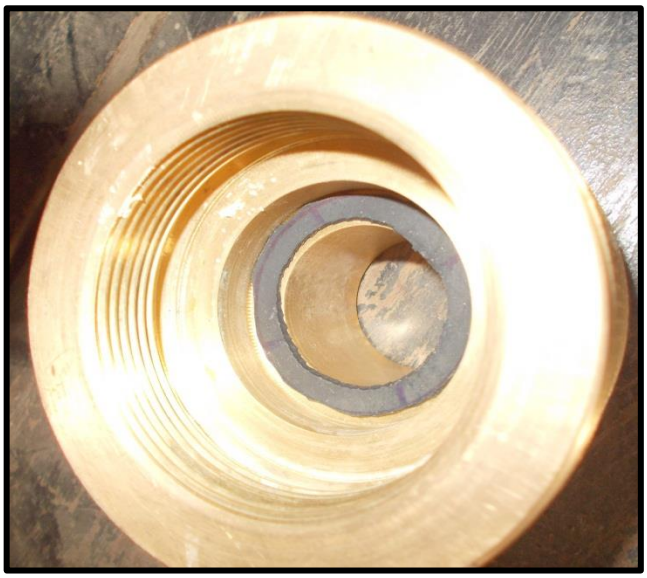

Fig. 4.2 O-ring inside the sample holder

Fig. 4.3 Sample inside the sample holder

Fig. 4.4 Variable size of sample holders

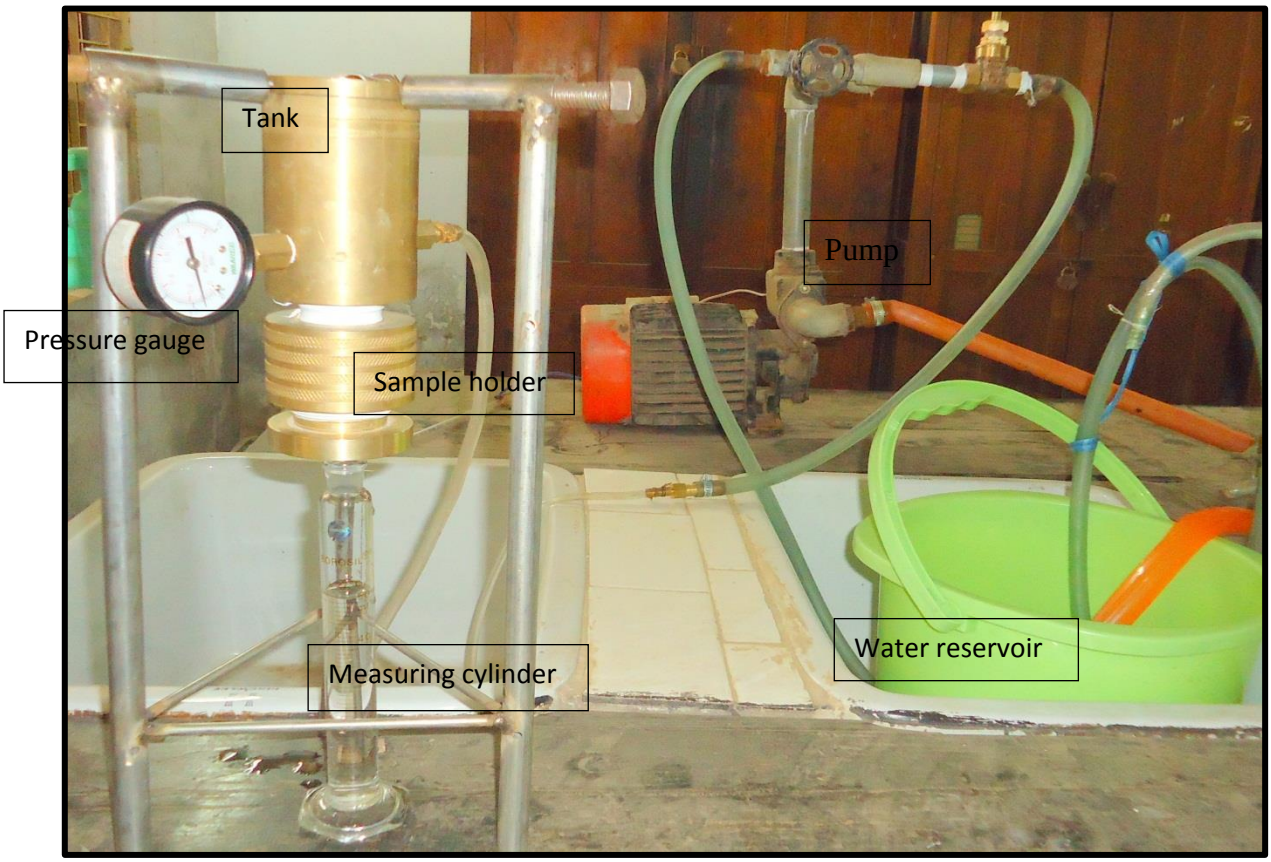

Fig. 4.5 Picture of complete permeability measurement set up

CHAPTER-5: PREPARATION OF POROUS SAMPLE

Porous sample was prepared by the following three methods

5.1 Sample Preparation by Pellet Pressing method:

5.1.1 Preparation of hydroxyapatite powder by precipitation method:

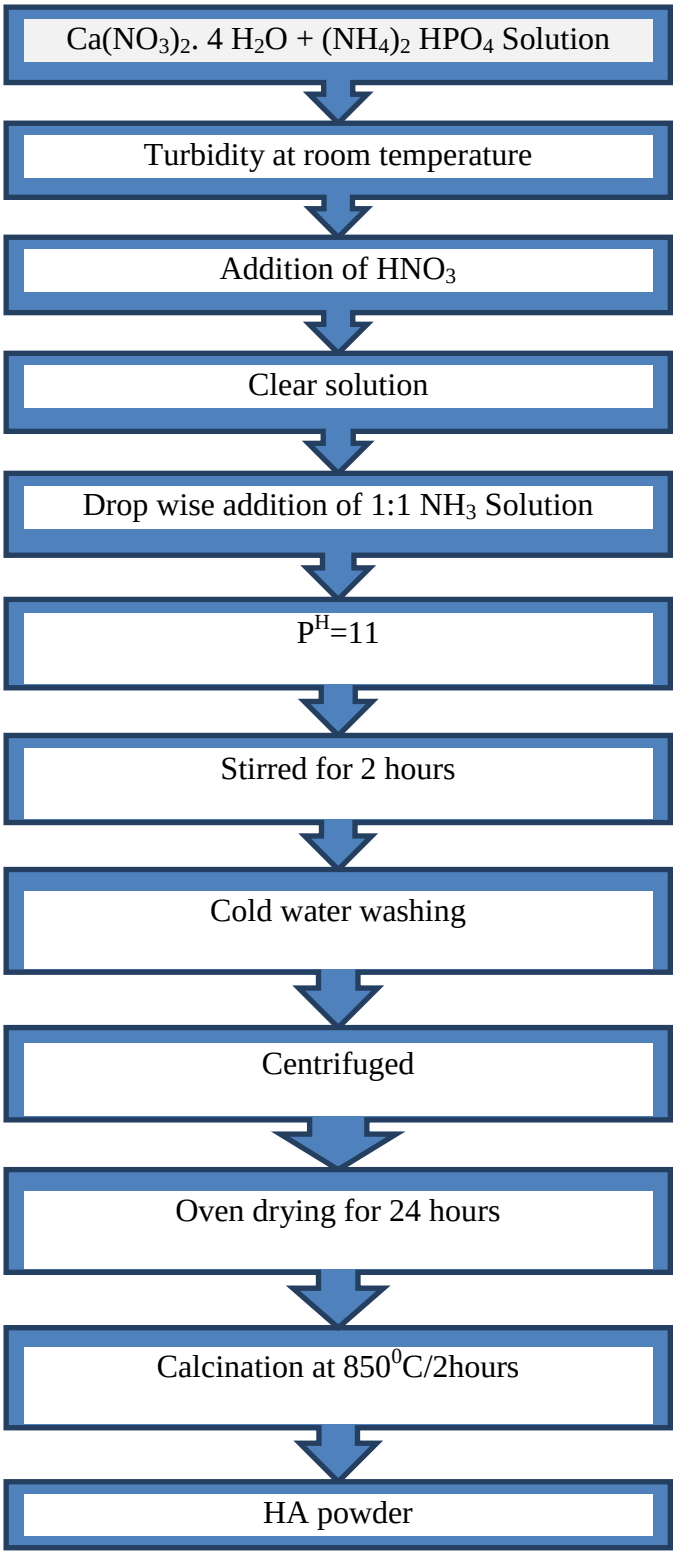

Fig. 5.1 Flow chart for HA powder preparation

5.1.2 Batch Preparation:

Dried powder (HA+ 5% PVA) was sieved through 100µm size sieve. Pore former naphthalene was first sieved through 200µm size sieve and the output was sieved with 100µm sieve to obtain naphthalene of -200µm+100µm. Batching was done by taking hydroxyapatite and naphthalene in the weight ratio 50:50, 60:40 and 70:30.

5.1.3 Pressing:

Green pellets were made by pressing in a hydraulic at a load of 3 tonnes for 90 second. Steel die with 25 mm diameter was used. The thickness of green pellet was 6 mm. Acetone was used for cleaning the die to avoid contamination and stearic acid solution was used for lubrication.

5.1.4 Drying:

Green pellet was dried at a temperature of 70-80^{0}C for about 24 hours.

5.1.5 Sintering of pellets:

The green pellets were sintered in an electrical resistance heating furnace at 1250^{0}C.The samples were held at 600^{0}C for 1 hour for binder removal and at the sintering temperature for 2 hours. The heating rate was at 3ºC/min till the final sintering temperature. The samples were cooled and taken out of the furnace after it cooled down to less than 100^{0}C.

5.2 Sample Preparation by Gel casting method:

Gel casting is a technique in which dispersed slurry is prepared with ceramic powder, binder, deflocculant and organic monomers. After the shaping and moulding of the slurry, it is allowed to gel. This technique can be used to prepare complex shapes. The disadvantage of the gel casting process is that high shrinkage is observed from gelling and subsequent demoulding stage [5-6].

For HA slurry preparation, calcined crystalline hydroxyapatite powder with a size of below 100µm was used as starting powder and Darvan C (Vanderbilt Organization Inc.), a25% aqueous solution of ammonium polymethacrylate, was used as a dispersant. The other components of the gelcasting process were organic monomers: acrylamide, $C_2H_3CONH_2$ and polymer methylene bisacrylamide $(C_2H_3CONH)_2CH_2$.Ammonium persulphate $(NH_4)_2S_2O_8$ and N,N,N,N tetramethylethylenediamine (TEMED) were used as initiator and catalyst respectively. Naphthalene (size -300+200µm) was dispersed with slow stirring. The flow diagram of the process is given below.

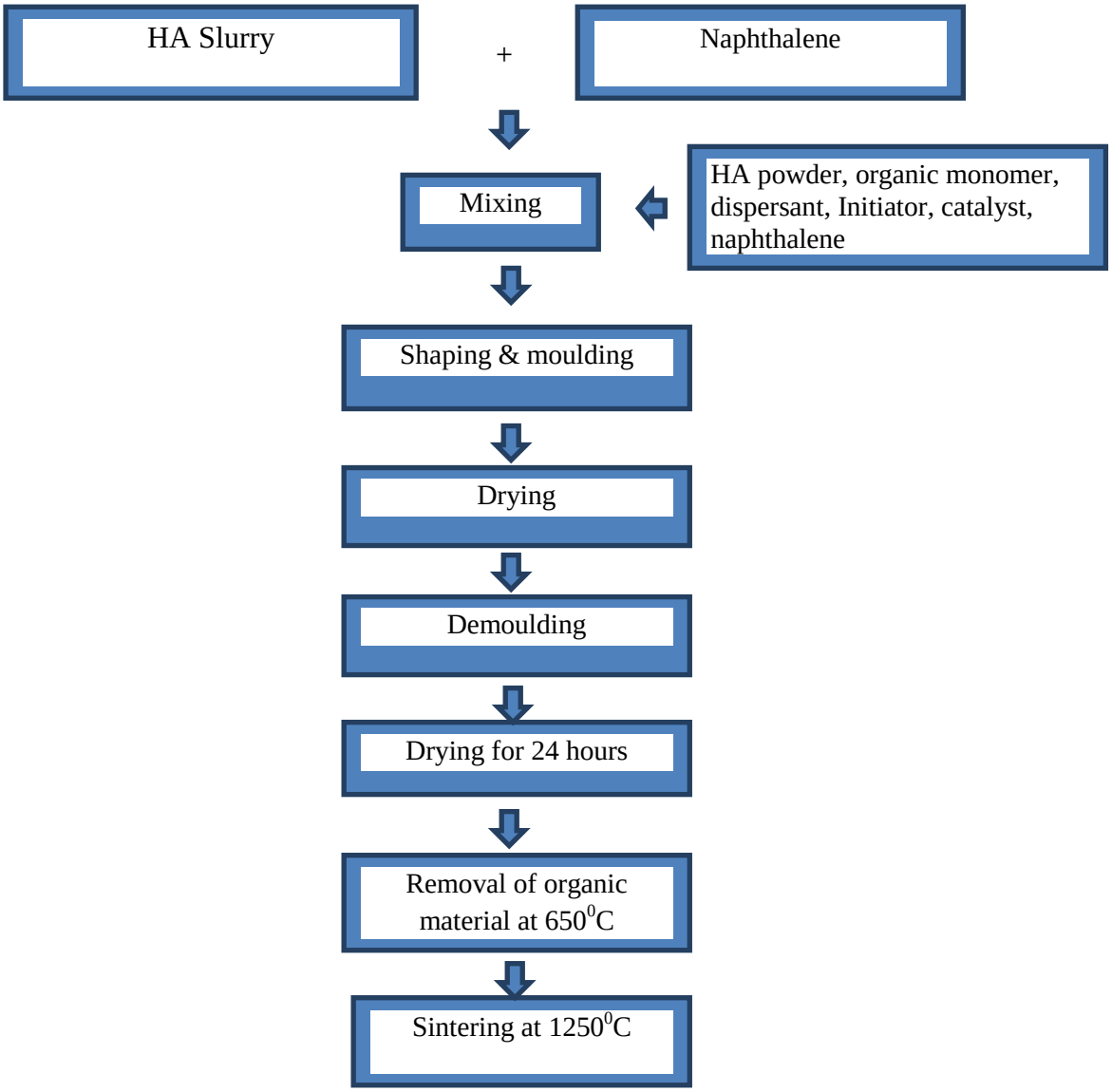

Fig.5.2 Process flow chart for gel casting sample

5.3 Sample Preparation by Polymer replication method:

This process involves repeated coating and dipping of a polymer sponge in ceramic slurry. During firing removal of inner polymer leads to formation of ceramic skeleton scaffold or porous structure. Porous structure with porosities varying from 70% to 90% can be prepared from this method [6-7].

5.3.1 Batch calculation:

Slurry with 50(vol. %) solid loading was prepared with following composition:

Reactive Alumina (Ceramic powder)

PVA (10%) (Binder)

Darvan C (2%) (Deflocculant)

Water (38%)

Density of alumina = 3.9 gm. /cc

Density of water = 1 gm. /cc

So for 50% solid loading amount of alumina required= 3.9 × 50 = 195 gm.

Water required = 1 × 50 = 50 ml

10% PVA solution= 10ml

2% deflocculant = 2 ml

So actual quantity of water required = 50 - (10+2) = 38ml

So final composition is:

Reactive alumina powder = 195 gm.

Amount = 38 ml

PVA solution = 10ml

Deflocculant = 2 ml

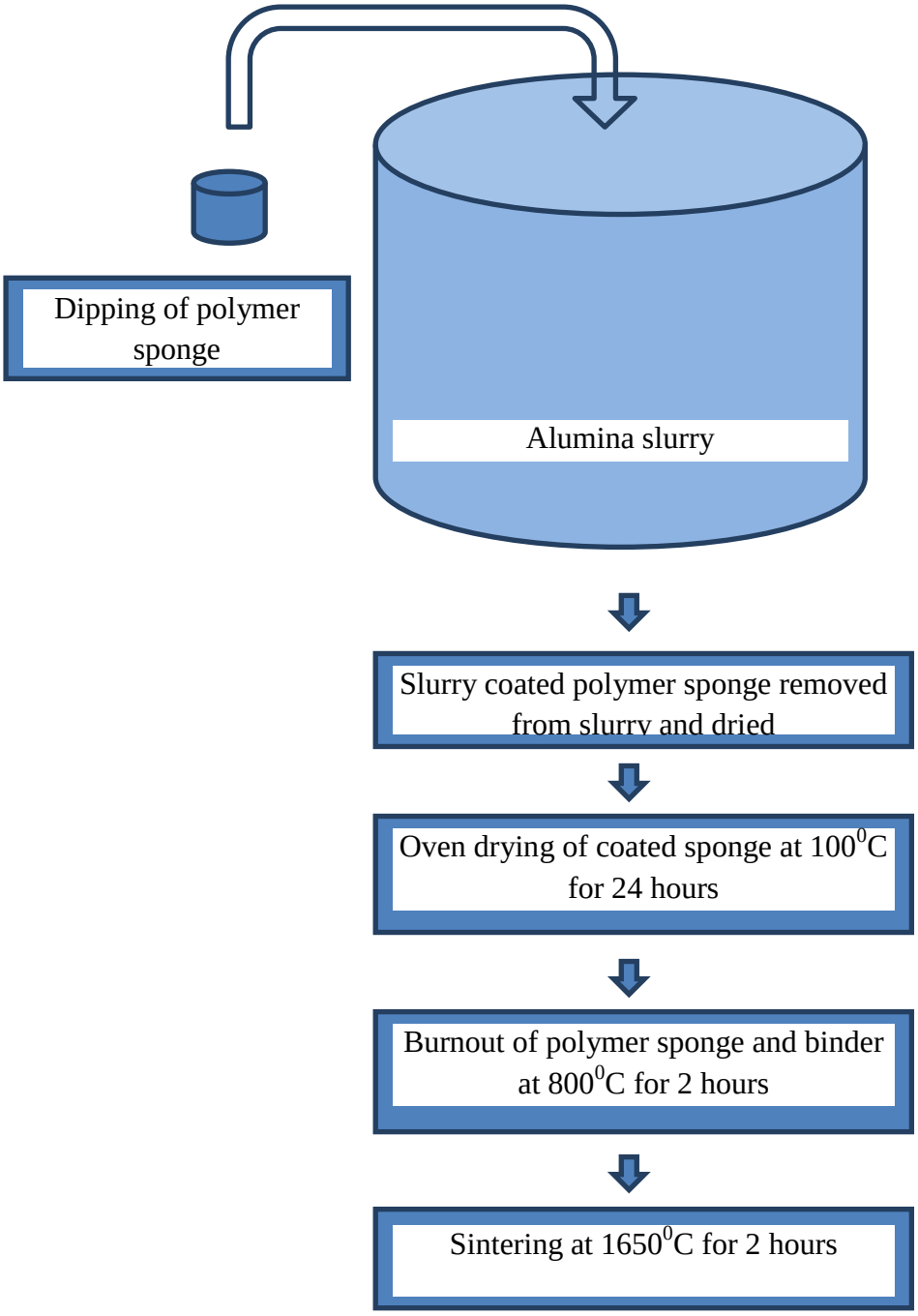

Fig. 5.3 Flow chart for polymer sponge replication method

5.3.2 Preparation of slurry:

In a beaker 38 ml water was taken and deflocculant Darvan C was added followed by stirring of solution by magnetic stirrer for 1 hour. Then PVA was added and again stirred for 30 minutes and finally alumina powder was added with continuous stirring for about 3 hours.

5.3.3 Cutting of polymer sponge and dipping:

Polymer sponge was cut according to the sample specification. Then cut sponge was dipped in the slurry for 5 times. The density of the sample is directly proportional to the number of dipping.

5.3.4 Drying:

After dipping, the sample was left for air drying for around 24 hours followed by oven drying in an electrical resistance heating oven at 90^0C to 100^0C for 24 hours.

5.3.5 Sintering:

Sintering of dried sample was done in an electrical resistance heating furnace at 1650^0C.At sintering temperature the holding time was for 2 hours and the intermediate soaking period of 2 hour at 800^0C for the burnout of polymer sponge. The heating rate was at 3ºC/min till 800ºC followed by heating at 5ºC/min till the final sintering temperature. The samples were held at the final temperature for 2 hours after which the furnace was switched off and furnace was allowed to cool. The samples were taken out of the furnace after temperature reached below 100ºC.

CHAPTER-6:
EXPERIMENTAL WORK

6.1 Apparent Porosity, Bulk Density Measurement:

The B.D and A.P of sintered sample was measured as per the following formula

B.D = D / (W-S)

A.P = ((W-D) × 100) / (W-S)

Where D- Dry weight

S- Suspended weight

W- Soak weight

The dry weight (D) of sample was measured. The weighed samples were soaked in water kept inside a beaker and were evacuated in a vacuum evacuator to allow all the air bubbles to come out from samples. The samples were kept in vacuum for further 30 minutes to allow escaping all the air bubbles. After removing from vacuum evacuator, the suspended weight(S) and soaked weight (W) of the samples were measured.

Table 6.1 Bulk density and apparent porosity of the sample prepared by different method.

Sample type	**Sample no.**	**Dry wt. (D) (gm.)**	**Suspended wt.(S) (gm.)**	**Soaked wt. (W) (gm.)**	**B.D= D/(W-S) (gm./cc)**	**A.P = (W-D)*100 /(W-S) (%)**
50% naphthalene pellet pressing sample	1.	3.48	2.48	5.65	1.09	68.48
	2.	3.29	2.32	5.23	1.13	66.63
40% naphthalene pellet pressing sample	1.	5.51	3.55	8.34	1.15	59.19
	2.	5.41	3.43	8.24	1.12	58.81
30% naphthalene pellet pressing sample	1.	6.67	4.33	9.65	1.25	55.97
	2.	6.48	4.18	9.47	1.22	56.57
Polymer sponge sample	1.	10.19	7.23	12.53	1.92	44.08
60% naphthalene gel casting sample	1.	9.21	6.94	16.06	1.01	75.10
50% naphthalene gel casting sample	1.	10.99	7.11	17.35	1.07	62.12

6.2 Measurement of Permeability:

Table 6.2 Specific permeability coefficient calculation for sample prepared by different method

Sample type	Time of flow (sec.)	Vol. of water passed (ml)	Area exposed (m^2)	Pressure given (Pa)	Thickness of the sample (m)	Specific permeability coefficient (m^2)
40% naphthalene pellet pressing sample	1200	3	2.27×10^{-4}	15×10^{4}	0.006	3.92×10^{-16}
30% naphthalene pellet pressing sample	1200	2	2.27×10^{-4}	15×10^{4}	0.006	2.61×10^{-16}
Polymer sponge sample	35.97	50	2.27×10^{-4}	1.9×10^{4}	0.01507	4.188×10^{-12}
Polymer sponge sample	18.06	47	2.27×10^{-4}	3.9×10^{4}	0.01507	3.918×10^{-12}
Polymer sponge sample	17.44	50	2.27×10^{-4}	4.9×10^{4}	0.01507	3.445×10^{-12}
Polymer sponge sample	15.25	48	2.27×10^{-4}	5.8×10^{4}	0.01507	3.159×10^{-12}
60% naphthalene gel casting sample	9.73	547	7.068×10^{-4}	5×10^{4}	0.019	2.68×10^{-11}
50% naphthalene gel casting sample	11	247	7.068×10^{-4}	5×10^{4}	0.014	7.8×10^{-7}

6.3 Effect of pressure on volume flow rate:

According to the Darcy's equation the volume flow rate is directly proportional to the pressure. Figure 6.1 (shown below) provides the volume flow rate increases with increasing pressure.

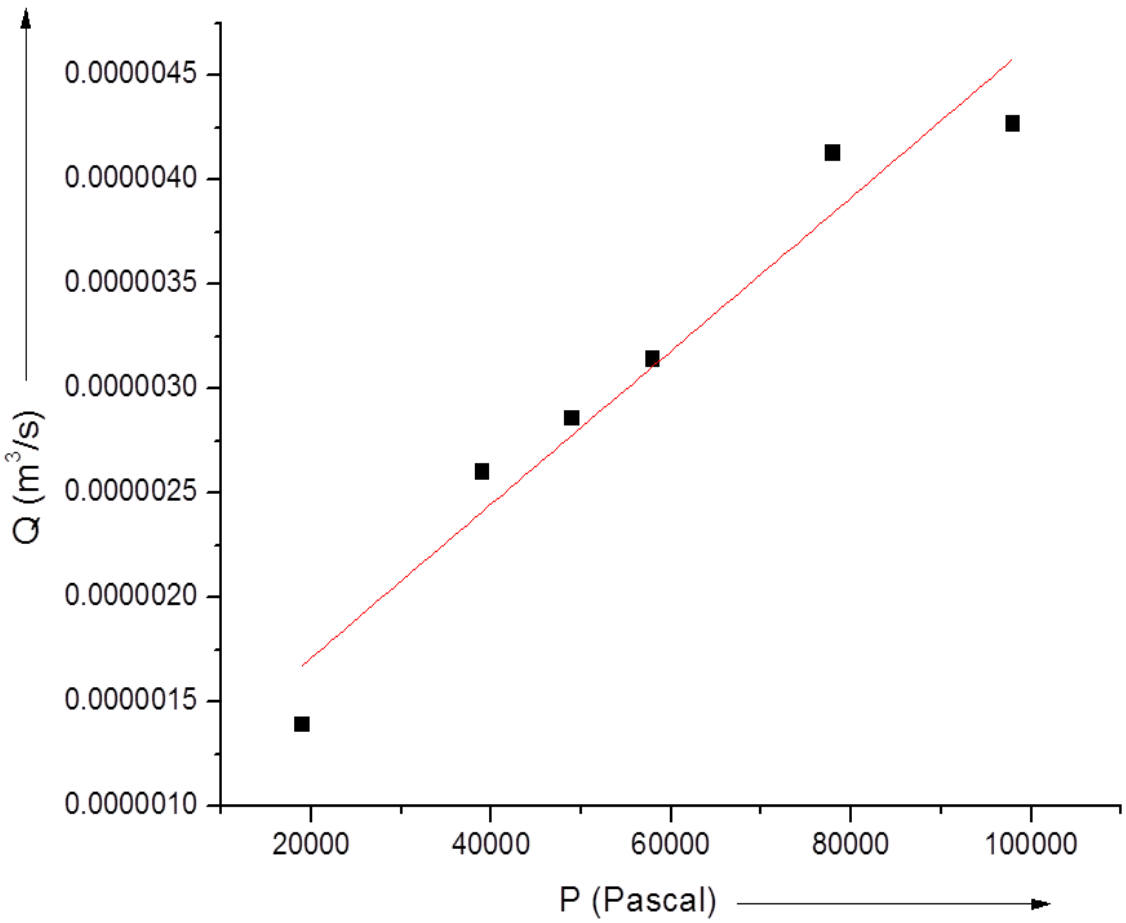

Fig. 6.1 Variation of volume flow rate with pressure

6.4Pore Size Distribution by Porosimetry Test:

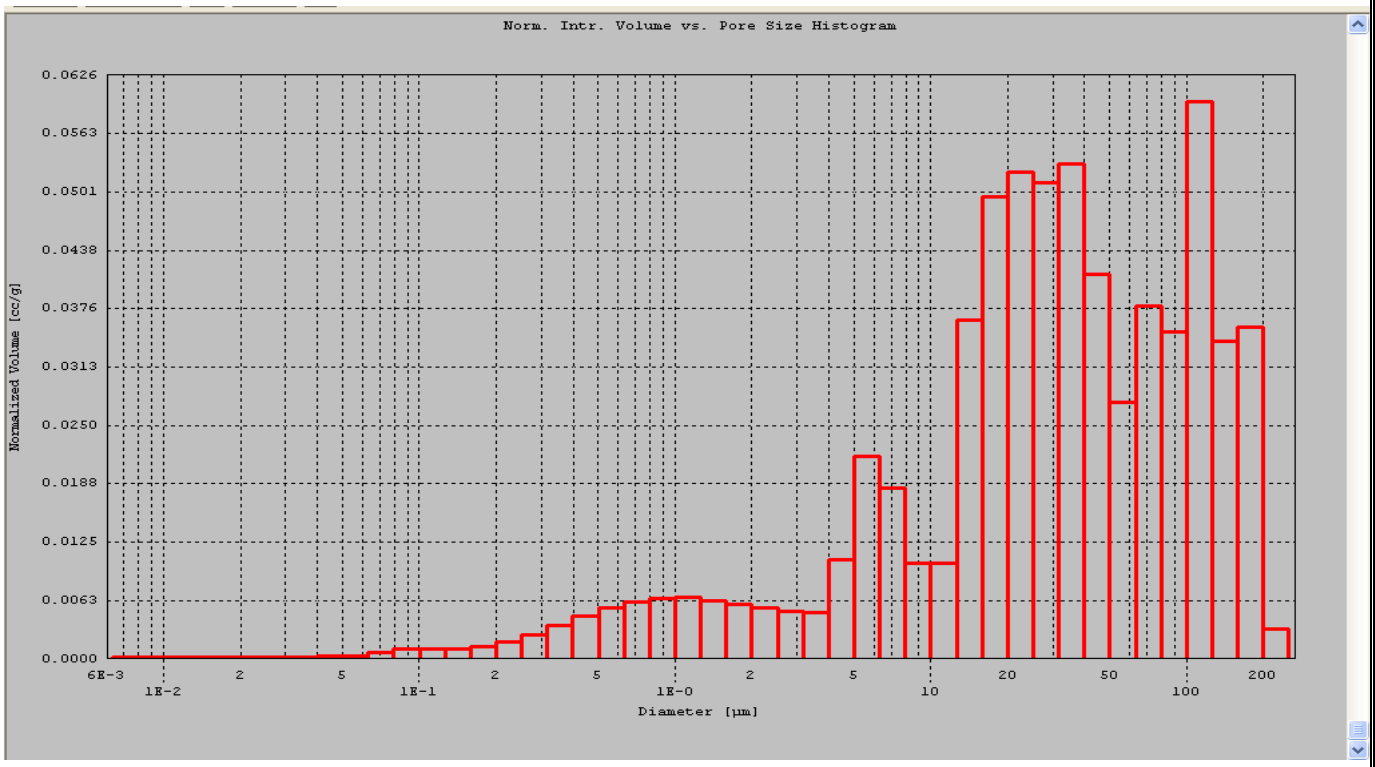

Fig.6.2 Pore size distribution of 60% naphthalene gel casting sample. (Pore diameter range 0.006719µm to 261.75µm).

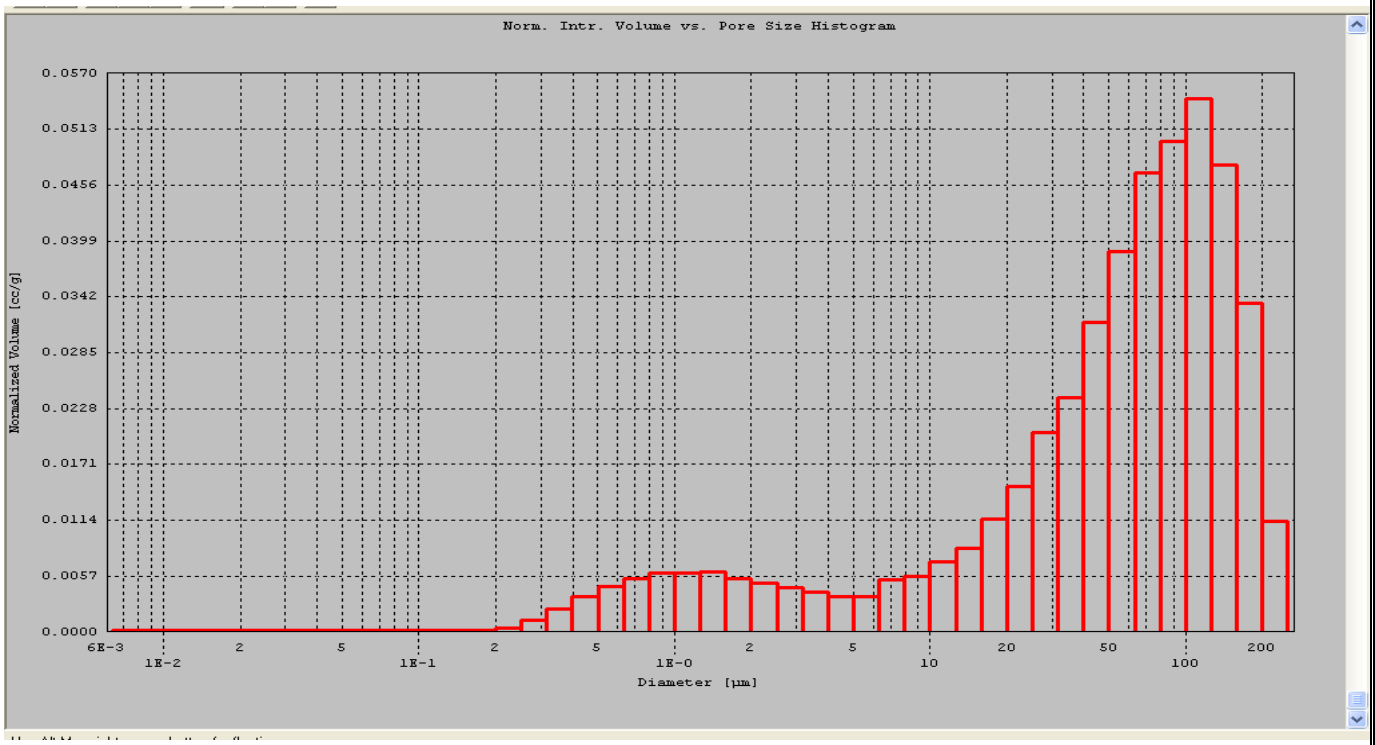

Fig. 6.3 Pore size distribution of 50% naphthalene gel casting sample.(Pore diameter range 0.00667µm to 230.268µm)

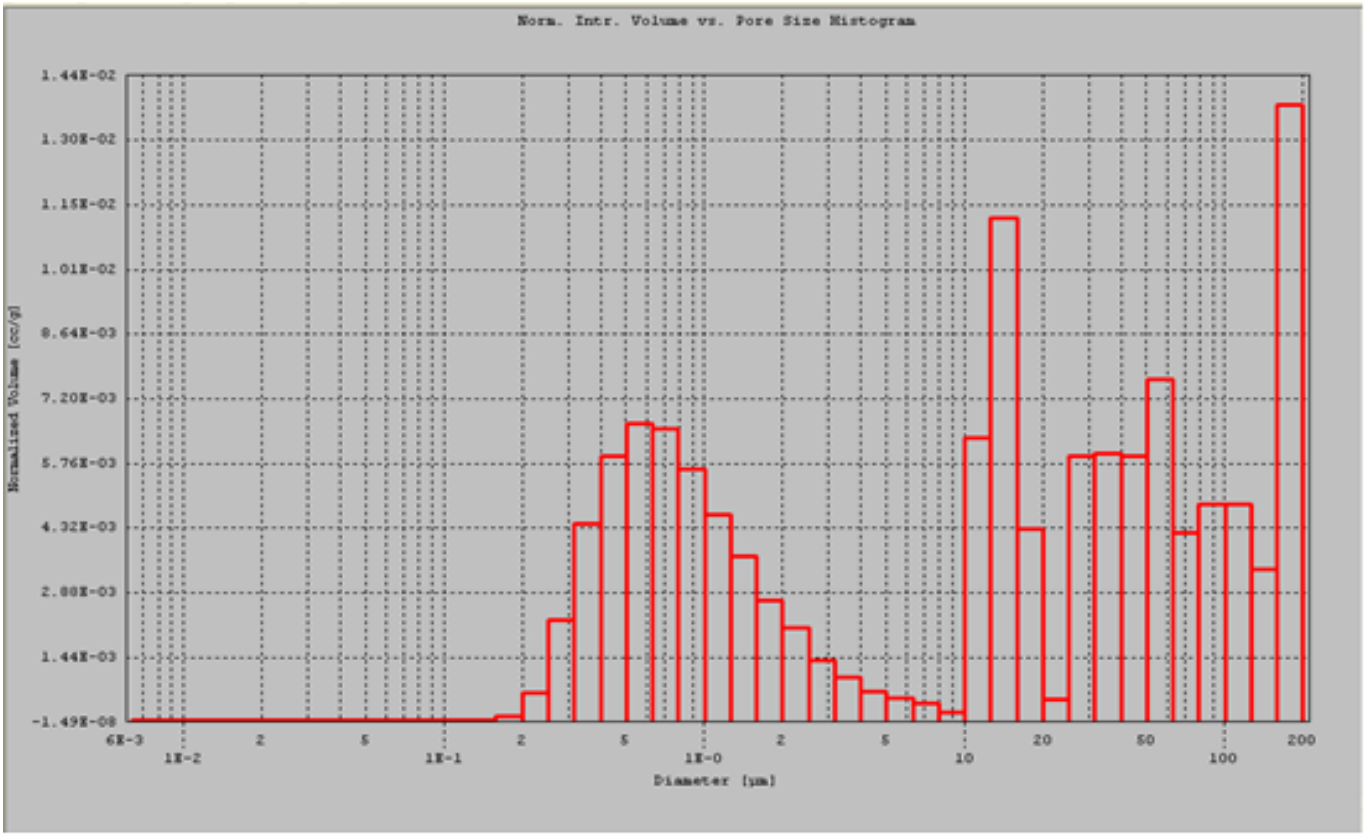

Fig.6.4 Pore size distribution of polymer sponge sample. (Pore diameter range 0.00672µm to 194.923µm)

6.5 Calculation:

Using **Lukasiewicz** equation [1]

$$\mathbf{Kp=\Phi R_L^{\,2}/K_L}$$

Where,

Kp- coefficient of specific permeability

Φ- vol. fraction porosity

R_L- vol. average linear mean pore radius determined by porosimetry

K_L-constant for a particular pore network. For cylindrical pore, K_L=8

Table 6.3 Summary of Apparent porosity, average pore radius and average pore diameter of sample prepared by different methods.

Sample	**Apparent porosity (%)**	**Average pore radius (R_L)(μm)**	**Average pore diameter. (μm)**
60% naphthalene gel casting sample	75.10	16.92	33.84
50% naphthalene gel casting sample	62.12	10.04	20.08
Polymer sponge sample	44.08	8.71	17.42

CHAPTER-7:

RESULTS &DISCUSSION

7.1 Results and Discussion:

The main aim of the study was to design, fabricate and test a permeability measurement apparatus. The second aim was to calibrate the apparatus with some standard sample and to correlate the permeability value obtained from the fabricated permeability equipment with that obtain from a commercially available equipment.

From the study following could be concluded

- A simple design for the permeability measurement apparatus could be prepared.
- The apparatus could be fabricated with the help of Central Workshop using the design.
- The apparatus was leak proof.
- Some porous samples were tested in the apparatus and the permeability value was recorded for these samples.
- Although no calibration was made because of lack of the standard sample but from the permeability value measured by the apparatus the following could be concluded that permeability depends (1) On the sample property (2) Flow rate vs. pressure show the linear relationship in accordance with the standard equation.
- Although apparatus was not calibrated the permeability of the different samples corresponded well with the data obtain from other system. For example a sample prepared by the pellet pressing method with naphthalene as pore former although had higher porosity did not show interconnected pores in the microstructure. Such a sample is expected to have low permeability and the result obtained from the fabricated apparatus exhibited similar trend. On the other hand sample prepared by gel casting method and polymer sponge replication method has shown high pore connectivity in the SEM microstructure. Such sample has also shown higher permeability measured by this apparatus.

Therefore it can be said that although the absolute value may become different if the instrument is calibrated, the present study showed that the apparatus can measure the permeability with relative ease and probably with accuracy. However the last part needs to verified with the standard samples having known porosity, permeability and pore size.

CHAPTER-8:
CONCLUSION

8.1 Conclusion:

A simple and low cost permeability measurement apparatus was designed and the permeability obtained from the fabricated apparatus show similar trend when measured by the other equipment (Mercury porosimetry). The equipment needed to be calibrated using standard sample with known permeability and porosity. Another interesting feature is that, this apparatus can be used for pressure casting provided that we put the slurry in a tubular mesh container and apply air pressure.

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